UCID- 17974

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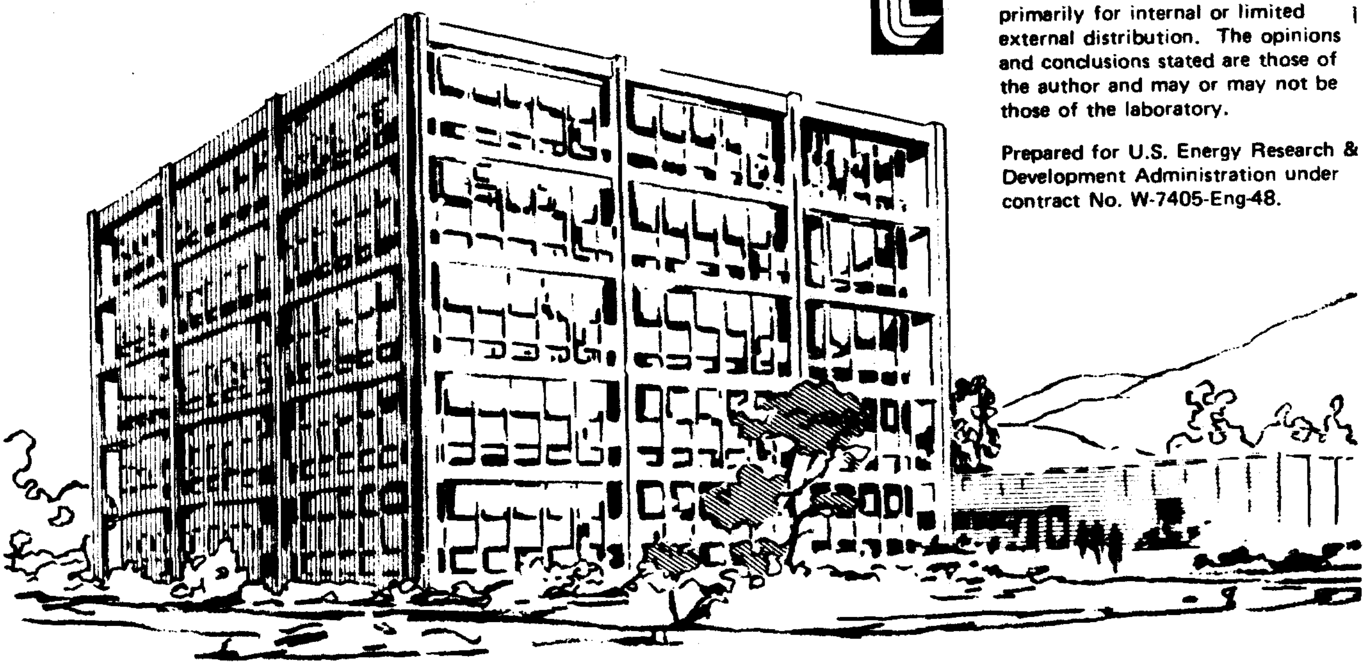
November, 1978



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Prepared for U.S. Energy Research & Development Administration under contract No. W-7405-Eng-48.

FLOW NON-UNIFORMITIES IN PACKED BEDS:

EFFECT ON DISPERSION

R.G. Carbonell

Department of Chemical Engineering

University of California

Davis, California 95616

September 1978

Abstract

A general equation is derived for the dispersion coefficient in a packed bed under conditions of non-uniform, one dimensional flow. Predictions of the theory are analyzed for the simple case of two concentric annular regions in a cylindrical packed bed with different void fractions. Small differences in void fraction can lead to significant changes in the dispersion coefficient for the packed bed, both in laminar and turbulent flow regimes. The radial variations of the average residence time and spread of a pulse of non-reactive tracer are investigated.

1. INTRODUCTION

The existence of radial void fraction distributions in packed beds of particles is well established. Benenate and Brosilow [1] report void fractions in packed beds of spheres approaching 1 near the wall and exhibiting a damped oscillation up to 9 particle diameters into the bulk of the bed where the void fraction is 0.4. Similar distributions have been measured by several other investigators [2,3,4]. Three very recent articles have focused attention on the importance of this wall effect on transport parameters in packed beds. Schlünder [5] and Martin [6] were able to account for the extremely low values of the Nusselt numbers measured experimentally in packed beds at particle Peclet numbers less than 10, by considering the void fraction in the packed bed as consisting of two separate regions. In a region at the center of the bed whose area is no less than 90% of the total bed area, the void fraction was considered constant at 0.40, while in the remainder of the bed, up to the wall, the void fraction was considered to have an average value of 0.50. The difference in fluid velocities in the two regimes was enough to account for orders of magnitude differences in the Nusselt number at low Peclet numbers due to the importance of channeling at the wall at these low flow rates. Botterill and Denlåye [7] used a function that would describe how the void fraction would vary as a function of distance away from a cylindrical rod imbedded in a packed bed of spheres in order to properly take into account the local flow velocity in their estimates of heat transfer coefficients from the rod to the spheres. Using this model they obtained much better agreement with the available data than was possible with models that considered the fluid velocity around the rods to be the same as the fluid velocity corresponding to the average bed void fraction. One is encouraged by the success of these investigators to explore the consequences of this void fraction distribution on dispersion in packed beds. This would improve our understanding of dispersion in general and possibly allow for better interpretations of dispersion coefficients in packed bed reactors. Lerou and Froment [8] and Schertz and Bischoff [9] measured velocity profiles in packed beds that clearly indicate regions of high flow rate near the wall, thus providing additional experimental evidence as to the existence of the void distribution and its importance in packed bed reactors.

Our approach here is to consider the case of one-dimensional flow in a packed bed subject to a void fraction distribution as described above. The non-uniformity in the void fraction will result in a non-uniform velocity profile that can be estimated from the Ergun equation [10]. Following the approach pioneered by Taylor [11], we obtain a general equation for the dispersion coefficient valid for any flow non-uniformity. As a specific example we consider the void fraction distribution chosen by Schlünder [5] and Martin [6] in their analysis. Computations using this model indicate that these flow non-uniformities can have a drastic effect on the dispersion coefficient for the packed bed. The response of the bed to pulses of inert tracer are investigated in order to see how the mean residence time and spread of the pulse would vary as a function of radial position.

2. THEORY

We begin our analysis by considering the mass transport equation for a non-reactive solute in a porous media in terms of the intrinsic phase-averaged concentration in the fluid phase $\langle c\rangle^{\alpha}$, and the intrinsic phase average velocity $\langle v\rangle^{\alpha}$,

$$\frac{\partial\langle c\rangle^{\alpha}}{\partial t} + \langle \mathbf{v}\rangle^{\alpha}\cdot\nabla\langle c\rangle^{\alpha} = \nabla\cdot(\mathbf{D}\cdot\nabla\langle c\rangle^{\alpha}) \tag{1}$$

The development of this equation has been considered in detail by Gray [12], Slattery [13] and Whitaker [14, 15], using volume-averaging techniques. The tensor $\mathbf{D}$ is a dispersion tensor that describes the local dispersion of the solute in the α phase anywhere in the bed. Of course, the components of $\mathbf{D}$ are functions of the local void fraction and local velocity in the α phase. In Eq. (1) we have assumed the solute does not diffuse into the solid particles in the bed, that it is found in low concentrations, and that the flow in the bed is incompressible. Since we are neglecting transport of solute into the particles, the results of this theory would apply to gas phase dispersion in a bed of non-porous particles such as oil shale rubble, and to liquid phase dispersion where the solute or tracer used in a dispersion experiment has a molecular diameter much greater than the largest pore diameter of the particles.

The intrinsic phase averaged velocity in the α phase may be computed by using the volume averaged continuity equation,

$$\nabla\cdot\langle \mathbf{v}\rangle^{\alpha} = 0 \tag{2}$$

together with Darcy's law in the creeping flow regime [16],

$$\langle \mathbf{v}\rangle^{\alpha} = -\frac{\mathbf{K}}{\varepsilon\mu}\cdot\nabla\langle P\rangle^{\alpha} \tag{3}$$

where $\underset{\approx}{K}$ is the permeability tensor. For cases where inertial terms may be of importance, a vectorial form of Ergun's equation [10] could yield a reasonable estimate of the flow distribution.

If we limit ourselves to the case of one dimensional flow in the z direction then Eqs. (2) and (3) reduce to,

$$\frac{d\langle v_z\rangle^{\alpha}}{dz} = 0 \tag{4}$$

and

$$\langle v_z\rangle^{\alpha} = -\frac{K_{zz}}{\varepsilon\mu}\frac{\partial\langle P\rangle^{\alpha}}{\partial z} \tag{5}$$

where K_{zz} and ε are considered to be only a function of position perpendicular to the flow and all off-diagonal elements of $\underset{\approx}{K}$ are taken to be zero. It is easy to show that the gradient in the pressure in the z direction is a constant,

$$\frac{\partial\langle P\rangle^{\alpha}}{\partial z} = -\left(\frac{\Delta P}{\ell}\right) \tag{6}$$

so that

$$\langle v_z\rangle^{\alpha} = \frac{K_{zz}}{\varepsilon\mu}\left(\frac{\Delta P}{\ell}\right) , \tag{7}$$

A good estimate of the z component of the permeability tensor may be obtained from the Blake-Kozeny equation [10] for laminar flow,

$$K_{zz} = \frac{d_p^{\,2}}{150}\,\frac{\varepsilon^3}{(1-\varepsilon)^2} , \tag{8}$$

where d_p is a particle diameter based on the specific surface to volume ratio a_v of the particle,

$$d_p = 6/a_v \tag{9}$$

For turbulent flow $\langle v_z\rangle^\alpha$ may be estimated from the Burke-Plummer equation [10],

$$\langle v_z\rangle^\alpha = \left(\frac{2d_p}{3.5\,\rho}\ \frac{\varepsilon^3}{(1-\varepsilon)}\right)^{\frac{1}{2}}\ \frac{1}{\varepsilon}\ \left(\frac{\Delta P}{\ell}\right)^{\frac{1}{2}} \tag{10}$$

In applying Eqs. (9) and (10), it is assumed that $\Delta P/\ell$ is constant in the direction perpendicular to flow. We see that if the void fraction ε varies even a slight amount as a function of position perpendicular to the flow, the fluid velocity will be greatly affected due to the strong dependence of $\langle v_z\rangle^\alpha$ on ε, specially in the laminar flow regime.

If we allow all off-diagonal elements of $\underset{\approx}{D}$ to be zero, and consider the ratio of the transverse to longitudinal dispersion coefficients to be a constant, then Eq. (1) for one-dimensional flow in cylindrical coordinates reduces to,

$$\frac{\partial\langle c\rangle^\alpha}{\partial t} + \langle v_z\rangle^\alpha\ \frac{\partial\langle c\rangle^\alpha}{\partial z} = \frac{\partial}{\partial z}\left(D_{zz}\ \frac{\partial\langle c\rangle^\alpha}{\partial z}\right) + \frac{1}{r}\ \frac{\partial}{\partial r}\left[r\left(\alpha D_{zz}\ \frac{\partial\langle c\rangle^\alpha}{\partial r}\right)\right] \tag{11}$$

where

$$\frac{D_{rr}}{D_{zz}} = \alpha \tag{12}$$

Greenkorn and Kessler [17] cite the results of various experimenters indicating that the ratio of longitudinal to transverse coefficients of dispersion (α^{-1}) lies in the range of 3 to 10 for unconsolidated porous media and can be as large as 60 for consolidated solids. This ratio remains constant with varying flow rate. Since the velocity $\langle v_z \rangle^\alpha$ is considered for one dimensional flow to be only a function of radial position, and since D_{zz} is a function of $\langle v_z \rangle^\alpha$ then, if we assume α is independent of flow rate Eq. (11) can be written as,

$$\frac{\partial c}{\partial t} + v_z(r)\,\frac{\partial c}{\partial z} = D\,\frac{\partial^2 c}{\partial z^2} + \frac{\alpha}{r}\frac{\partial}{\partial r}\left(r\,D\,\frac{\partial c}{\partial r}\right) \tag{13}$$

where for simplicity we have dropped the phase average symbol from the concentration and the velocity, and it is understood that D is the axial component of the dispersion tensor. We can make Eq. (13) dimensionless by defining the following dimensionless variables,

$$\begin{aligned} &C = c/c_o, \quad Z = z\mathcal{D}/u_oR^2, \quad \eta = r/R, \quad U_z(\eta) = v_z(r)/u_o \\ &D^* = D/\mathcal{D}, \quad \theta = t\mathcal{D}/R^2, \quad P_e = u_oR/\mathcal{D} \end{aligned} \tag{14}$$

where R is the bed radius, $\mathcal{D}$ is the molecular diffusivity of the solute, u_o is a characteristic velocity, usually taken as the area averaged interstitial velocity over the bed cross section and c_o is some reference concentration, usually chosen to be the maximum concentration of an input peak for a pulse input. Using these definitions, Eq. (13) becomes

$$\frac{\partial C}{\partial \theta} + U_z(\eta)\,\frac{\partial C}{\partial Z} = \frac{D^*}{P_e^{\,2}}\,\frac{\partial^2 C}{\partial Z^2} + \frac{\alpha}{\eta}\frac{\partial}{\partial \eta}\left(\eta\,D^*\frac{\partial C}{\partial \eta}\right) \tag{15}$$

Now we consider the case of a pulse of tracer introduced into a semi-infinite packed bed at time $\theta = 0$, so that boundary and initial conditions for Eq. (16) can be written as,

$$\frac{\partial C}{\partial \eta} = 0, \quad \eta = 0 \text{ and } \eta = 1 \tag{16}$$

$$C = \delta(\theta), \quad Z = 0 \tag{17}$$

$$\frac{\partial C}{\partial Z} = 0 \qquad Z \to \infty \tag{18}$$

We would like to develop an equation for the area-averaged concentration in the fluid, namely, for the quantity,

$$\langle C \rangle = 2 \int_0^1 C\ (\eta, Z, \theta)\eta d\eta \tag{19}$$

We can multiply Eq. (15) by 2η, integrate from 0 to 1 and apply the boundary conditions (16) to obtain,

$$\frac{\partial \langle C \rangle}{\partial \theta} + \frac{\partial}{\partial Z} \langle U_z C \rangle = \frac{1}{P_e^{\,2}} \frac{\partial^2}{\partial Z^2} \langle D^* C \rangle \tag{20}$$

where the brackets denote the integral defined by Eq. (19). We now express U_z, C and D* in terms of their area-averaged values and derivations from the area-averages [12],

$$C = \langle C \rangle + \tilde{C} \tag{21}$$

$$U_z = \langle U_z \rangle + \tilde{U}_z \tag{22}$$

$$D^* = \langle D^* \rangle + \tilde{D}^* \tag{23}$$

Substituting Eqs. (21)-(23) into Eq. (20) and making use of the property of the deviations

$$\langle\tilde{C}\rangle = \langle\tilde{U}_z\rangle = \langle\tilde{D}^*\rangle = 0 \tag{24}$$

we obtain

$$\frac{\partial\langle C\rangle}{\partial\theta} + \langle U_z\rangle \frac{\partial\langle C\rangle}{\partial Z} = \frac{\langle D^*\rangle}{P_e^2} \frac{\partial^2\langle C\rangle}{\partial Z^2} - \frac{\partial}{\partial Z}\langle\tilde{U}_z\tilde{C}\rangle + \frac{\partial^2}{\partial Z^2}\langle\tilde{D}^*\tilde{C}\rangle \tag{25}$$

This is the dispersion equation for the packed bed in terms of the area-averaged interstitial velocity and the area-averaged concentration. The first term on the right hand side contains a dispersion coefficient which is the area-averaged value of the local dispersion coefficients in the bed. The second term is the additional contribution to dispersion caused by velocity deviations from plug flow. The quantity $\langle\tilde{U}_z\tilde{C}\rangle$ is the axial component of the dispersion vector, and its properties have been discussed in detail by Whitaker [14]. The last term on the right hand side is a new term that arises from deviations in the local dispersion coefficients. As we shall show later, this term will contribute to the skewness of the pulse, and under some conditions may be shown to be smaller than the other terms in the equation. What we need to do now is to obtain an expression for $\tilde{C}$, in terms of $\langle C\rangle$ and calculate $\langle\tilde{U}_z\tilde{C}\rangle$ and $\langle\tilde{D}^*\tilde{C}\rangle$. This will be accomplished by applying ideas originally developed by Taylor [11] for studying dispersion in laminar flow in a tube. The development presented here is formally different from his approach, and we will be able to obtain criteria for when the approximations made can be expected to be valid.

We begin by transforming Eq. (15) to a coordinate system $\bar{Z}$ moving with the velocity of the pulse U_p which we assume to be unknown,

$$\bar{Z} = Z - U_p\theta \tag{26}$$

so that Eq. (15) becomes

$$\frac{\partial C}{\partial \theta} + \left[U_z(\eta) - U_p\right]\frac{\partial C}{\partial \bar{Z}} = \frac{D^*}{P_e^2}\frac{\partial^2 C}{\partial \bar{Z}^2} + \frac{\alpha}{\eta}\frac{\partial}{\partial \eta}\left(\eta D^* \frac{\partial C}{\partial \eta}\right) \tag{27}$$

Looking at the order of magnitudes of the terms in this equation, it is clear that for times θ such that

$$\left[\alpha\, \theta <D^*>\right] >> 1 \tag{28}$$

the time derivative will be much smaller than the term having to do with dispersion in the radial direction. Similarly, dispersion in the axial direction may be neglected relative to convection in the axial direction if

$$\left[Pe^2\, L<U_z>/<D^*>\right] >> 1 \tag{29}$$

where L is a dimensionless characteristic pulse length. As time becomes larger and larger both criterion (28) and (29) will be easier to meet since the pulse length L increases with θ. If both (28) and (29) are satisfied, a psuedo-steady state approximation may be invoked on Eq. (15) so that it reduces to the form,

$$\left[U_z(\eta) - U_p\right] \frac{\partial C}{\partial \bar{Z}} = \frac{\alpha}{\eta} \frac{\partial}{\partial \eta} \left(\eta\, D^* \frac{\partial C}{\partial \eta}\right) \tag{30}$$

Using the definition (21) for $\tilde{C}$ and making the additional assumption (used successfully by Taylor [11]),

$$\frac{\partial \langle C \rangle}{\partial \bar{Z}} \gg \frac{\partial \tilde{C}}{\partial \bar{Z}}$$

we obtain a second order ordinary differential equation for $\tilde{C}$,

$$\left[U_z(\eta) - U_p\right] \frac{\partial \langle C \rangle}{\partial \bar{Z}} = \frac{\alpha}{\eta} \frac{d}{d\eta} \left(\eta\, D^* \frac{\partial \tilde{C}}{\partial \eta}\right) \tag{31}$$

with boundary conditions,

$$\frac{\partial \tilde{C}}{\partial \eta} = 0 \quad \text{at} \quad \eta = 0, 1 \tag{32}$$

and the additional constraint (24).

Integrating (31) once and using the boundary condition at $\eta = 0$, one obtains

$$\frac{d\tilde{C}}{d\eta} = \left\{ \frac{1}{\alpha D^*} \frac{1}{\eta} \int_0^{\eta} \left[U_z(\eta') - U_p\right] \eta'\, d\eta' \right\} \frac{\partial \langle C \rangle}{\partial \bar{Z}} \tag{33}$$

When we apply boundary condition (32) at $\eta = 1$ we find immediately that

$$U_p = 2 \int_0^1 U_z(\eta')\eta' d\eta' = \langle U_z \rangle \tag{34}$$

indicating that the pulse will travel at the average interstitial velocity of the fluid. This being the case, we can write Eq. (34) in the form,

$$\frac{d\tilde{C}}{d\eta} = \left\{ \frac{1}{\alpha D^*} \ \frac{1}{\eta} \int_0^\eta \tilde{U}_z(\eta')\eta' d\eta' \right\} \frac{\partial \langle C \rangle}{\partial \bar{Z}} \tag{35}$$

where use has been made of Eq. (22). Integrating (35), we find an expression for $\tilde{C}$,

$$\tilde{C} = A + \left\{ \alpha^{-1} \int_0^\eta \frac{d\xi}{D^*(\xi)\xi} \int_0^\xi \tilde{U}_z(\eta')\eta' d\eta' \right\} \frac{\partial \langle C \rangle}{\partial \bar{Z}} \tag{36}$$

The unknown coefficient A may be evaluated by the condition $\langle \tilde{C} \rangle = 0$. If we label the double integral in Eq. (36), $\Phi(\eta)$, then imposing this constraint results in the final expression for $\tilde{C}$,

$$\tilde{C} = \alpha^{-1} \left[\Phi(\eta) - 2 \int_0^1 \Phi(\eta)\eta \, d\eta \right] \frac{\partial \langle C \rangle}{\partial \bar{Z}} = F(\eta) \frac{\partial \langle C \rangle}{\partial \bar{Z}} \tag{37}$$

showing, as is expected [14], that the deviation $\tilde{C}$ is proportional to the gradient of $\langle C \rangle$ in the $\bar{Z}$ direction, the direction of flow. Evaluating the area integrals,

$$\langle \tilde{U}_z \tilde{C} \rangle = \langle \tilde{U}_z F(\eta) \rangle \frac{\partial \langle C \rangle}{\partial \bar{Z}} , \tag{38}$$

$$\langle \tilde{D}^* \tilde{C} \rangle = \langle \tilde{D}^* F(\eta) \rangle \frac{\partial \langle C \rangle}{\partial \bar{Z}} , \tag{39}$$

and substituting the result into Eq. (23), we obtain the final form for the dispersion equation.

$$\frac{\partial \langle C\rangle}{\partial \theta} + \langle U_z\rangle \frac{\partial \langle C\rangle}{\partial Z} = \left[\frac{\langle D^*\rangle}{P_e^{\,2}} - \langle \tilde{U}_z F(\eta)\rangle\right] \frac{\partial^2 \langle C\rangle}{\partial Z^2} + \langle \tilde{D}^* F(\eta)\rangle \frac{\partial^3 \langle C\rangle}{\partial Z^3} \qquad (40)$$

where $F(\eta)$ is defined by Eq. (37). The term multiplying the second derivative is the dispersion coefficient for the packed bed. The last term on the right hand side contributes to the skewness of the pulse.

Once the integrals in $\Phi(\eta)$ and $F(\eta)$ have been evaluated, it is a simple matter to calculate specific expressions for the coefficients in the dispersion Eq. (40). As a specific example, we consider the flow in the packed bed as described by two concentric annular regions of different velocities, the same model used by Schlünder [5] and Martin [6] in their analysis of heat transfer data.

3. DISPERSION IN ANNULAR FLOW

Consider the interstitial fluid velocity and local dispersion coefficients in the packed bed to be described by the functions,

$$U_z(\eta) = \begin{cases} U_0 = v_0/u_0 & 0 < \eta < \xi \\ U_1 = v_1/u_0 & \xi < \eta < 1 \end{cases} \qquad (41)$$

$$D^*(\eta) = \begin{cases} D_0^* = D_0/\mathcal{D} & 0 < \eta < \xi \\ D_1^* = D_1/\mathcal{D} & \xi < \eta < 1 \end{cases} \qquad (42)$$

where ξ is some fraction of the total bed radius. The area averaged velocity and dispersion coefficients are easily shown to be,

$$\langle U_z\rangle = U_o\,\xi^2 + U_1(1-\xi^2) \tag{43}$$

$$\langle D^*\rangle = D_o^*\,\xi^2 + D_1^*(1-\xi^2) \tag{44}$$

so that the deviations of these quantities from their area-averaged values are,

$$\tilde{U}_z(\eta) = \begin{cases} -(U_1 - U_o)(1-\xi^2) & 0 < \eta < \xi \\ (U_1 - U_o)\,\xi^2 & \xi < \eta < 1 \end{cases} \tag{45}$$

$$\tilde{D}^*(\eta) = \begin{cases} -(D_1^* - D_o^*)(1-\xi^2) & 0 < \eta < \xi \\ (D_1^* - D_o^*)\,\xi^2 & \xi < \eta < 1 \end{cases} \tag{46}$$

Equation (30) then takes the form,

$$\begin{aligned} -\frac{(U_1 - U_o)(1-\xi^2)}{\alpha D_o^*}\,\frac{\partial\langle C\rangle}{\partial\bar{Z}} &= \frac{1}{\eta}\,\frac{d}{d\eta}\left(\eta\,\frac{d\tilde{C}}{d\eta}\right) \quad 0 < \eta < \xi \\ \frac{(U_1 - U_o)\xi^2}{\alpha D_o^*}\,\frac{\partial\langle C\rangle}{\partial\bar{Z}} &= \frac{1}{\eta}\,\frac{d}{d\eta}\left(\eta\,\frac{d\tilde{C}}{d\eta}\right) \quad \xi < \eta < 1 \end{aligned} \tag{47}$$

If we define the parameters,

$$A = \frac{\Delta U(1-\xi^2)}{\alpha D_o^*}\,\frac{\partial\langle C\rangle}{\partial\bar{Z}} \quad , \tag{48}$$

$$B = \frac{\Delta U\,\xi^2}{\alpha D_1^*}\,\frac{\partial\langle C\rangle}{\partial\bar{Z}} \quad , \tag{49}$$

where

$$\Delta U = U_1 - U_o \, , \tag{50}$$

then Eq. (31) becomes

$$-A = \frac{1}{\eta} \frac{d}{d\eta}\left(\eta \frac{d\tilde{C}}{d\eta}\right) \qquad 0 < \eta < \xi \tag{51}$$

$$B = \frac{1}{\eta} \frac{d}{d\eta}\left(\eta \frac{d\tilde{C}}{d\eta}\right) \qquad \xi < \eta < 1 \tag{52}$$

with boundary conditions,

$$\frac{d\tilde{C}}{d\eta} = 0 \qquad \eta = 0, 1 \tag{53}$$

$$\tilde{C} \text{ continuous at } \eta = \xi \tag{54}$$

Again we require that $\langle\tilde{C}\rangle = 0$.

Solving Eqs. (51) and (52) subject to these constraints we obtain,

$$\tilde{C} = (A + B)\left(\frac{\xi^2}{4} - \frac{\xi^4}{8}\right) + B\left(\frac{\xi^2}{4} - \frac{3}{8} - \frac{1}{2}\ln\xi\right) - A\frac{\eta^2}{4} \qquad 0 < \eta < \xi \tag{55}$$

$$\tilde{C} = B\left(\frac{\xi^2}{4} - \frac{3}{8}\right) - (A + B)\frac{\xi^4}{8} + B\left(\frac{\eta^2}{4} - \frac{1}{2}\ln\eta\right) \qquad \xi < \eta < 1 \tag{56}$$

By combining (55) and (56) with (45) and (46) we can perform the appropriate area averages to calculate the coefficients in the dispersion equation,

$$<\tilde{U}_z\tilde{C}> = -\frac{(\Delta U)^2}{\alpha}\left\{\frac{\xi^4(1-\xi^2)^2}{8D_o^*} + \frac{[4\xi^6 - \xi^4\,(3 + 4\,\ell n\,\xi) - \xi^8]}{8D_1^*}\right\}\frac{\partial<C>}{\partial\bar{Z}} \tag{57}$$

$$<\tilde{D}^*\tilde{C}> = -\frac{\Delta D^*\Delta U}{\alpha}\left\{\frac{\xi^4(1-\xi^2)^2}{8D_o^*}\quad\frac{[4\xi^6 - \xi^4\,(3 + 4\,\ell n\,\xi) - \xi^8]}{8D_1^*}\right\}\frac{\partial<C>}{\partial\bar{Z}} \tag{58}$$

where $\Delta D^* = D_1^* - D_o^*$ (59)

It is convenient to write the term ΔU in terms of ratios of Peclet numbers,

$$\Delta U = U_1 - U_o = \frac{\Delta Pe}{Pe} = \frac{v_1 - v_o}{u_o} \tag{60}$$

where $\Delta Pe = (v_1 - v_o)R/\mathcal{D}$ (61)

Substituting (57) and (58) into the dispersion Eq. (25) we obtain,

$$\frac{\partial<C>}{\partial\theta} + <U_z>\frac{\partial<C>}{\partial Z} = \frac{K^*}{P_e^2}\frac{\partial^2<C>}{\partial Z^2} - \frac{S^*}{Pe}\frac{\partial^3<C>}{\partial Z^3} \tag{62}$$

where

$$K^* = <D>^* + \lambda^*, \tag{63}$$

$$\lambda^* = \frac{(\Delta Pe)^2}{\alpha}\left\{\frac{\xi^4(1-\xi^2)^2}{8D_o^*} + \frac{[4\xi^6 - \xi^4\,(3 + 4\ell n\,\xi) - \xi^8]}{8D_1^*}\right\}, \tag{64}$$

and

$$S^* = \frac{(\Delta D^*)(\Delta Pe)}{\alpha}\left\{\frac{\xi^4(1-\xi^2)^2}{8D_o^*} + \frac{[4\xi^6 - \xi^4(3 + 4\ell n\xi) - \xi^8]}{8D_1^*}\right\} \tag{65}$$

Notice that the dispersion coefficient K^* in Eq. (62) has two contributions, one from the area average of the local dispersion coefficients $\langle D\rangle^*$ while the second, λ^*, is the effect of the velocity distribution in the packed bed to dispersion. The term S^* represents the contribution of the velocity profile in the bed to the skewness of the pulse. From a simple order of magnitude estimate of the terms on the right hand side of Eq. (62) we see that the second derivative term will dominate the term with the third derivative if the condition,

$$\frac{K^*}{S^*}\frac{L}{Pe} \gg 1 \tag{66}$$

is satisfied.

Now we calculate the order of magnitude of the terms in Eq. (62) by considering both laminar and turbulent flow regimes. Following the example of Schlünder [5], we consider the central core of the packed bed where the velocity is v_o as comprising 90% of the total area, so that $\xi^2 = 0.9$, and $\xi = 0.949$. From the data cited by Greenkorn and Kessler [17], we pick a value of the ratio of the longitudinal to transverse dispersion coefficient equal to 6.5 so that it is somewhere between 3 and 10 for unconsolidated porous media. This makes $\alpha = 0.154$. Again following Schlünder [5], we let $\varepsilon_o = 0.40$ at the bulk of the bed and $\varepsilon_1 = 0.50$ at the wall. In laminar flow in the packed bed, Eqs. (7) and (8) predict that the ratio of the <u>interstitial</u> velocities will be,

$$\frac{v_1}{v_o} = \left(\frac{\varepsilon_1}{\varepsilon_o}\right)^2\left(\frac{1-\varepsilon_o}{1-\varepsilon_1}\right)^2 = 2.25 \tag{67}$$

while for turbulent flow, Eq. (10) predicts a ratio,

$$\frac{v_1}{v_o} = \left[\frac{\varepsilon_1}{\varepsilon_o}\left(\frac{1-\varepsilon_o}{1-\varepsilon_1}\right)\right]^{1/2} = 1.225 \tag{68}$$

We can now express the area-averaged interstitial velocity across the packed bed in terms of v_o. For laminar flow,

$$u_o = v_o\xi^2 + v_1(1-\xi^2) = 1.125\ v_o \tag{69}$$

while for turbulent flow,

$$u_o = 1.0225\ v_o \tag{70}$$

The ratio $\Delta Pe/Pe$ is equal to

$$\frac{\Delta Pe}{Pe} = \frac{v_1 - v_o}{u_o} = 1.11 \tag{71}$$

for laminar flow and

$$\frac{\Delta Pe}{Pe} = 0.22 \tag{72}$$

for turbulent flow. The only parameters we need to estimate now are the values of D_o^* and D_1^*. Bear [18] has summarized the results of many experimenters and presents a comprehensive plot of $D^* = Dzz/\mathcal{D}$ as a function of particle Peclet number, $Pe_p = \varepsilon\langle v_z\rangle^\alpha d_p/\mathcal{D}$. The results may be roughly summarized for our purposes as follows,

$$D^* \simeq 0.6 \qquad Pe_p < 1.0 \tag{73}$$
$$D^* \simeq Pe_p \qquad Pe_p > 1.0$$

These relationships obviously are not exact, but will be of great help to us in estimating the values of D_o^* and D_1^*. We divide the calculations into laminar and turbulent regimes. For laminar flow we let $Pe_p < 1.0$ everywhere in the bed so that $D_o^* = D_1^* = 0.6$ and the dispersion coefficient in Eq. (55) becomes,

$$\frac{K^*}{Pe^2} = \frac{0.6}{Pe^2} + \left|\frac{\Delta Pe}{Pe}\right|^2 \frac{1}{\alpha(0.6)}\left\{\frac{\xi^4(1-\xi^2)^2}{8} + \frac{[4\xi^6 - \xi^4(3 + 4\ell n\xi) - \xi^8]}{8}\right\} \tag{74}$$

$$= \frac{0.6}{Pe^2} + 1.45 \times 10^{-2}$$

where use has been made of Eqs. (63) and (64), and the estimate of $\Delta Pe/Pe$ for laminar flow. Clearly, for large values of the Peclet number, say for $Pe > 25$, the velocity dependent term will dominate so that the value of $K^* \simeq 1.45 \times 10^{-2} Pe^2$. It should be remembered that $Pe = Pe_p(R/\varepsilon d_p)$ so that if, for example, $Pe_p \simeq 0.5$, in order for the Peclet number to be as large as 25 then the ratio of the bed diameter to the particle diameter should be roughly equal to 20. This is easily satisfied for many industrial scale packed beds. Even when $Pe = 10$, the contribution from the effect of the non-uniform velocity profile will be of the same order as the contribution from the area-average of the local dispersion coefficients. Note that $S^* = 0$ for the case $Pe_p < 1.0$ since it depends on the difference $\Delta D^* = D_1^* - D_o^*$. The approximation (73) makes the second term on the right hand size of Eq. (62) identically equal to zero. For turbulent flow, Eq. (73) indicates that approximately,

$$\frac{D_1^*}{D_o^*} = \frac{\varepsilon_1 v_1}{\varepsilon_o v_o} = 1.53 \tag{75}$$

since $v_1/v_o = 1.225$ for the turbulent flow case. Equations (63) and (64) then combine to give,

$$\frac{K^*}{Pe^2} = \frac{D_o^*(1.053)}{Pe^2} + \left(\frac{\Delta Pe}{Pe}\right)^2 \frac{1}{\alpha D_o^*} \left\{ \frac{\xi^4(1-\xi^2)^2}{8} + \frac{[4\xi^6 - \xi^4(\xi + 4\ell n\xi) - \xi^8]}{(1.53)8} \right\} \tag{76}$$

where, according to (67) we can approximate D_o^* by,

$$D_o^* = (Pe_p)_o \tag{77}$$

Eq. (76) for turbulent flow then becomes,

$$\frac{K^*}{Pe^2} = \frac{1.053(Pe_p)_o}{Pe^2} + \frac{3.182 \times 10^{-4}}{(Pe_p)_o} \tag{78}$$

where the estimate of $\Delta Pe/Pe = 0.22$ for turbulent flow has been used.

Equation (78) indicates that unless $Pe > 57.5\ (Pe_p)_o$, the second term, which is the contribution to dispersion from the non uniform velocity, will be insignificant. Since the ratio,

$$\frac{Pe}{(Pe_p)_o} = \frac{1}{\varepsilon_o}\left(\frac{u_o}{v_o}\right)\left(\frac{R}{d_p}\right) = 2.56\left(\frac{R}{d_p}\right) \tag{79}$$

for turbulent flow, then this corresponds to a ratio $(R/d_p \simeq 23$. We can conclude then that in turbulent flow, if $R/d_p \ll 23$, the wall effect will dominate dispersion, while if R/d_p is significantly less than 23, the wall effect will be unimportant. It must be remembered that these results are obtained for a fixed ξ. Obviously as the ratio d_p/R varies one would expect ξ to change and significantly affect these conclusions.

An estimate of the coefficient multiplying the third derivative in Eq. (62) for turbulent flows yields,

$$\frac{S^*}{Pe} \simeq \frac{\Delta D^*}{\alpha D_o^*} \frac{\Delta Pe}{Pe} (1.0125 \times 10^{-3}) = 7.67 \times 10^{-4} \qquad (80)$$

where use has been made of Eqs. (75) and (72). The criterion (66) for this term being negligible in comparison to the dispersion term becomes, upon substituting Eq. (80),

$$(1.3 \times 10^3)\ K^* \frac{L}{Pe^2} \gg 1 \qquad (81)$$

When the second term in Eq. (78) is unimportant, Eq. (81) indicates that

$$(Pe_p)_o \frac{L}{Pe^2} \gg 7.3 \times 10^{-4} \qquad (82)$$

while when the second term dominates,

$$\frac{L}{(Pe_p)_o} \gg 2.4 \qquad (83)$$

The time when these criteria are satisfied of course will depend on individual values of Pe and $(Pe_p)_o$, however, it is not unreasonable to expect conditions under which the third derivative term in Eq. (62) could be neglected relative to the dispersion term under turbulent flow conditions.

4. Flow Non-uniformities and Pulse Responses

In some applications, it may be of interest to measure flow non-uniformities in a packed bed by inserting a pulse of inert tracer and measuring the differences in the residence times in the bed as a function of radial position. The formulation presented in Section II allows for a clear interpretation of the results of such an experiment. If we combine Eqs. (37) and (21), we obtain an equation for the point concentration in the bed, namely,

$$C(\eta,\theta,Z) = \langle C\rangle(\theta,Z) + F(\eta)\frac{\partial\langle C\rangle}{\partial Z} \tag{84}$$

in a stationary frame. An equation of the type (62) would be solved for $\langle C\rangle$ that would be introduced into Eq. (84),

$$\frac{\partial\langle C\rangle}{\partial\theta} + \frac{\partial\langle C\rangle}{\partial Z} = \frac{K^*}{Pe^2}\frac{\partial^2\langle C\rangle}{\partial Z^2} \tag{85}$$

where we have let $\langle U_z\rangle = 1.0$ and we have dropped the skewness term in accordance with the conclusions of Section 3. The boundary conditions for Eq. (85) would be,

$$\begin{aligned} \langle C\rangle &= f(\theta) \quad & Z &= 0 \\ \langle C\rangle &= 0 \quad & Z &\to \infty \\ \langle C\rangle &= 0 \quad & \theta &= 0 \end{aligned} \tag{86}$$

where $f(\theta)$ is the shape of the input pulse.

We define the moments of $\langle C\rangle$ with respect to time as,

$$M_k = \int_0^\infty \theta^k \langle C\rangle \, d\theta, \quad k = 0, 1, 2 \tag{87}$$

and the moments of the point concentration C with respect to time as,

$$m_k = \int_0^\infty \theta^k \, C(\eta,\theta,Z) \, d\theta \tag{88}$$

We can relate the m_k to the M_k by multiplying Eq. (84) by θ^k and integrating from 0 to ∞, to obtain,

$$m_o = M_o + F(\eta) \frac{dM_o}{dZ}, \tag{89}$$

$$m_1 = M_1 + F(\eta) \frac{dM_1}{dZ}, \tag{90}$$

and

$$m_2 = M_2 + F(\eta) \frac{dM_2}{dZ} \tag{91}$$

All we need to do now is to find M_o, M_1 and M_2. This can be accomplished by taking the Laplace Transform of Eq. (85) and the boundary conditions (86) with respect to time to obtain,

$$\overline{\langle c\rangle}(s) = \overline{f}(s) \exp[\beta(s)Z] \tag{92}$$

where

$$\beta(s) = \frac{Pe^2}{2K^*} \left[1 - \sqrt{1 + \frac{4K^*s}{Pe^2}}\right] \tag{93}$$

One can now use a well known theorem relating moments [19] to derivatives with respect to the Laplace Transform variable,

$$M_k = (-1)^k \lim_{s \to 0} \frac{d^k \overline{\langle c\rangle}(s)}{ds^k} \tag{94}$$

and find immediately,

$$M_o = \bar{f}(0), \tag{95}$$

$$M_1 = ZM_o + M_1(0), \tag{96}$$

$$M_2 = \frac{2K^*Z\,M_o}{Pe^2} + Z^2M_o + 2\,ZM_1(0) + M_2(0), \tag{97}$$

where the $M_k(0)$ are values of the moments at $\theta = 0$. Substituting Eqs. (95) - (97) into (89) - (91) and defining the absolute moments,

$$(\mu'_k)_\ell = m_k/m_o \tag{98}$$

$$(\mu'_k)_a = M_k/M_o \tag{99}$$

we obtain,

$$m_o = M_o \tag{100}$$

$$(\mu_1')_\ell = (\mu_1')_a + F(\eta) \tag{101}$$

$$(\mu_2')_\ell = (\mu_2')_a + 2F(\eta)\left[\frac{K^*}{Pe^2} + (\mu_1')_a\right] \tag{102}$$

Equation (101) is of particular interest to us since it predicts that the residence time at any radial position η is related to the residence time of the area-averaged concentration directly by the function $F(\eta)$. Thus, to measure $F(\eta)$ experimentally, one has to place a concentration probe at various positions along the radius, measure the outgoing pulses, calculate the average residence time at the individual points and subtract from this value the average residence time for the area-averaged concentration. For the annular flow case solved in Section III one can easily see that from Eqs. (55) and (56) that

$$F(\eta) = \alpha^{-1}\left(\frac{\Delta Pe}{Pe}\right)\left\{\frac{(1-\xi^2)}{D_o^*}\left[\frac{\xi^2}{4} - \frac{\xi^4}{8} - \frac{\eta^2}{4}\right] + \frac{\xi^2}{D_1^*}\left[\frac{\xi^2}{2} - \frac{\xi^4}{8} - \frac{3}{8} - \frac{1}{2}\ell n\xi\right]\right\} \tag{103}$$

for $0 < \eta < \xi$ and

$$F(\eta) = \alpha^{-1}\left(\frac{\Delta Pe}{Pe}\right)\left\{\frac{(1-\xi^2)}{D_o^*}\left(-\frac{\xi^4}{8}\right) + \frac{\xi^2}{D_1^*}\left[\frac{\xi^2}{4} - \frac{3}{8} - \frac{\xi^4}{8} + \frac{\eta^2}{4} - \frac{1}{2}\ell n\eta\right]\right\} \tag{104}$$

for $\xi < \eta < 1$.

In Figure 1, we show $F(\eta)$ for the laminar flow parameters, namely, $\alpha = 0.154$, $\Delta Pe/Pe = 1.11$, $D_o^* = D_1^* = 0.6$, and $\xi^2 = 0.90$. Even though the velocity profile has a discontinuity at $\eta = 0.949$, the effect on the residence times gets transmitted radially throughout the entire cross section. When $F(\eta)$ goes through zero, near $\eta = 0.7$, the pulse at that point has the same residence time as the area-averaged concentration.

Since the second central moment,

$$(\mu_2)_a = \frac{1}{m_o}\int_0^\infty [\theta - (\mu_1')_a]^2 \langle c\rangle \, d\theta \tag{105}$$

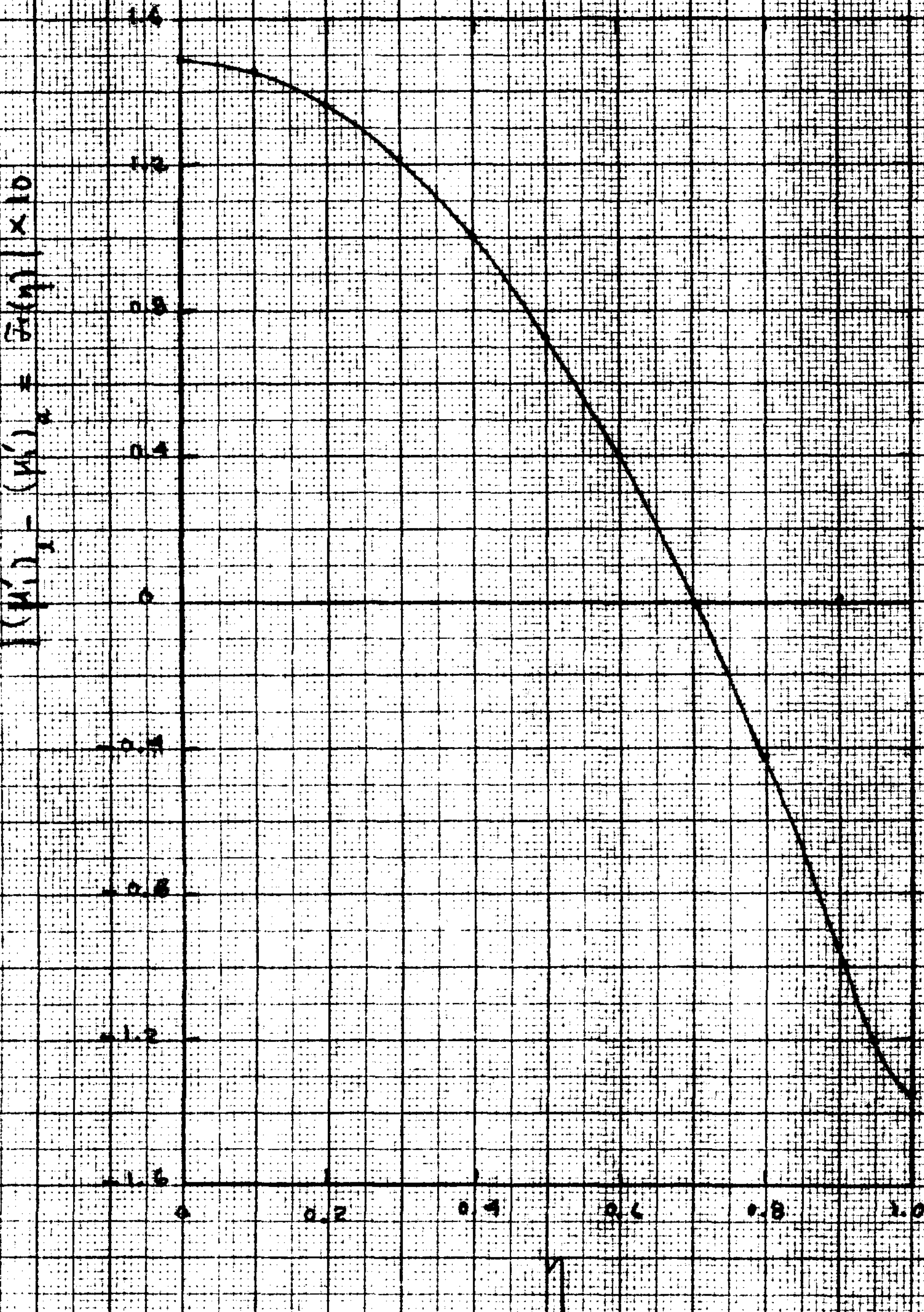
is related to the second absolute moment $(\mu_1')_a$ by the equation,

$$(\mu_2)_a = (\mu_2')_a - (\mu_1')_a^2 \tag{106}$$

it is not hard to show using Eq. (81) that

Figure 1

$(\mu_1')_1 - (\mu_1')_2$ for the laminar flow case.



$$(\mu_2)_\ell = (\mu_2)_a + \frac{2K^*}{Pe^2} \; F(\eta) - [F(\eta)]^2 \qquad (107)$$

where $(\mu_2)_\ell$ is the second central moment of the pulse output at a given radial position. The second central moment represents the spread of the pulse in time relative to its mean residence time. For laminar flow K^*/Pe^2 is given by Eq. (74) so that with a given fixed value of Pe we can calculate $(\mu_2)_\ell - (\mu_2)_a$ according to Eq. (107), (83) and (84). For the case $Pe = 50$ and laminar flow, the same conditions as the results obtained in Figure 1, Figure 2 shows how the spread of the pulses at given radial positions are related to the spread of the area averaged concentration. Equation (107) predicts that this quantity will be less than zero almost throughout the entire range of η, and have the largest negative value near the wall and at the center of the bed. The results of Figures 1 and 2 indicate possible ways in which flow maldistributions can be detected from measurements of pulse responses as a function of radial position in the bed.

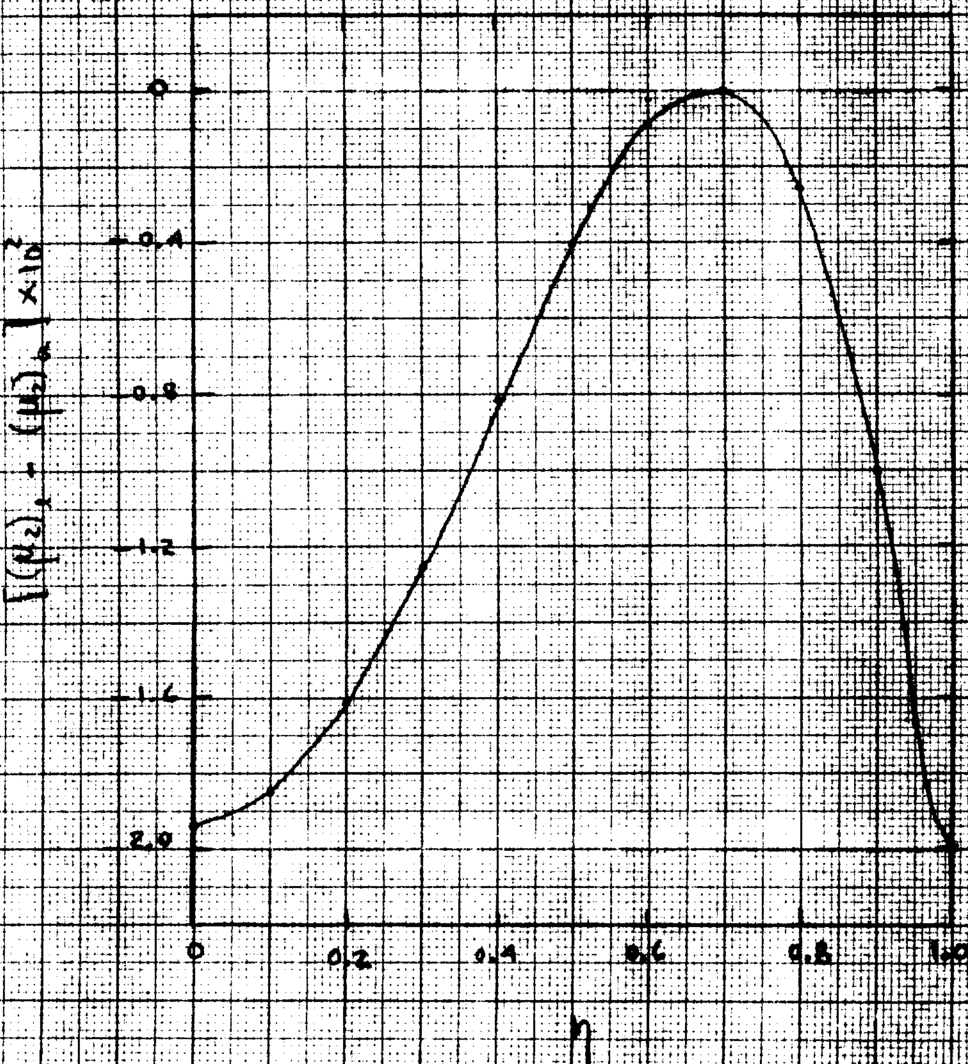
5. Conclusions

A general equation has been derived for the coefficients in the dispersion equation for the case of one-dimensional non-uniform flow in a packed bed. As an example, the implications of the model of Schlünder [5] and Martin [6] for flow non-uniformities in a packed bed have been investigated. Possible techniques for measuring the effect of flow non-uniformities have been developed by considering the response of the system to a pulse of non-reactive tracer.

Figure 2

$(\mu_2)_t - (\mu_2)_a$ for

laminar flow, $Pe = 50$.



Acknowledgement

The author wishes to thank Dr. R. Lyczkowski of Lawrence Livermore Laboratories for pointing out useful references related to this research, and for fruitful discussions of its results.

NOTATION

a_v	surface area per unit volume for particles
A	coefficient of integration
$\langle c\rangle^{\alpha}$, c	intrinsic phase-averaged solute concentration
c_o	characteristic concentration
C	dimensionless intrinsic phase-averaged concentration
$\langle C\rangle$	area-averaged value of C over the bed cross section
$\tilde{C}$	deviation of C from its area-averaged value
d_p	mean particle diameter
D, D_{zz}	axial component of the dispersion tensor
D_{rr}	radial component of the dispersion tensor
$\underset{\approx}{D}$	dispersion tensor
D^*	dimensionless axial component of the dispersion tensor
D_o^*, D_1^*	values of D^* for $0 < \eta < \xi$ and $\xi < \eta < 1$ respectively
ΔD^*	difference $D_1^* - D_o^*$
$\langle D^*\rangle$	area-averaged value of D^* over the bed cross section
$\tilde{D}^*$	deviation of D^* from its area-averaged value
$\mathcal{D}$	molecular diffusivity of the solute
$f(\theta)$	shape of the input pulse
$F(\eta)$	function multiplying the gradient in expression for $\tilde{C}$
K_{zz}	axial component of the permeability tensor

$\underset{\sim}{K}$	permeability tensor
K^*	dimensionless dispersion coefficient in dispersion equation for the bed
ℓ	length of bed
L	dimensionless pulse length
m_k	kth moment with respect to time of C
M_k	kth moment with respect to time of $\langle C \rangle$
$M_k(0)$	kth moment of $\langle C \rangle$ at $\theta = 0$
ΔP	pressure drop
$\langle P \rangle^\alpha$	intrinsic phase-averaged pressure
Pe	Peclet number for the bed, $U_o R/\mathcal{D}$
Pe_p	particle Peclet number, $\varepsilon \langle v_z \rangle^\alpha dp/\mathcal{D}$
$(Pe_p)_o$	particle Peclet number when $0 < \eta < \xi$
ΔPe	difference in Peclet numbers, $(v_1 - v_o) R/\mathcal{D}$
r	radial coordinate
R	bed radius
s	Laplace Transform variable
S^*	skewness parameter
t	time
u_o	area-averaged value of v_z over the bed cross section
U_z	dimensionless interstitial velocity in the axial direction
U_1, U_o	values of U_z from $0 < \eta < \xi$ and $\xi < \eta < 1$ respectively
ΔU	difference $U_1 - U_o$
U_p	dimensionless pulse velocity
$\langle U_z \rangle$	area-averaged value of U_z over the bed cross section
$\tilde{U}_z$	deviation of U_z from its area-averaged value

$\langle \underset{\sim}{v} \rangle^{\alpha}$	intrinsic phase-averaged velocity vector
$\langle v_z \rangle^{\alpha}$, v_z	axial component of the intrinsic phase-averaged velocity
v_o, v_1	interstitial velocities for $0 < \eta < \xi$ and $\xi < \eta < 1$ respectively
z	axial coordinate
Z	dimensionless axial coordinate
$\bar{Z}$	dimensionless axial coordinate moving at pulse velocity

Greek Symbols

α	ratio of transverse to longitudinal dispersion coefficients
ε	bed void fraction
ε_o,ε_1	void fraction for $0< \eta < \xi$ and $\xi < \eta < 1$ respectively
ξ	fraction of bed radius
η	dimensionless radial coordinate
θ	dimensionless time
$\beta(s)$	function of the Laplace Transform variable
$(\mu_k')_\ell$	kth absolute moment of C at a given η, Z
$(\mu_k')_a$	kth absolute moment of $\langle C \rangle$ at a given Z
$(\mu_k)_\ell$	kth central moment of C at a given η, Z
$(\mu_k)_a$	kth central moment of $\langle C \rangle$ at a given Z

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